Transverse effective dispersion coefficients in a chemically heterogeneous medium with flow fluctuations.

V. ZAVALA-SANCHEZ, M. DENTZ & X. SANCHEZ-VILA
Department of Geotechnical Engineering and Geosciences, Technical University of Catalonia (UPC), Z.P. 08034, Barcelona, Spain
e-mail: vanessa.zavala@upc.edu
e-mail: marco.dentz@upc.edu
e-mail: xavier.sanchez-vila@upc.edu

Abstract We investigate the effective transport of a sorptive solute through a chemically heterogeneous medium subject to temporal fluctuations of the flow conditions. We focus on linear instantaneous sorption, with a spatially variable retardation factor. The medium is physically homogeneous. The temporal variability of the flow is represented by a stationary random process. In this stochastic modelling framework, solute spreading is quantified by effective dispersion coefficients. Focusing on the transverse contributions due to chemical heterogeneity and temporal fluctuations, we find enhanced transverse spreading characterized by a transverse effective dispersion coefficient that, in contrast to transport in steady state flow, evolves to a disorder-induced macroscopic value independent of local dispersion.

Key words Stochastic Modelling, Effective Dispersion, Random Adsorption, Temporal Random Flow.

INTRODUCTION

Local scale physical and chemical medium heterogeneities lead to an effective large scale transport behaviour that is qualitatively and quantitatively different from the one observed in homogeneous media. The interaction of spatial fluctuations of the system parameters and local scale transport processes lead in general to enhanced solute spreading and mixing.

Attinger et al., 1999, and *Dentz et al.*, 2000a, 2000b, studied the temporal behaviour of dispersion coefficients in a stochastic modelling framework developed up to the second order in the fluctuations of the random fields, for a chemically and physically heterogeneous medium respectively under steady state flow conditions. Physical and chemical medium heterogeneities lead to an increase of the longitudinal dispersion, D_L, to a macroscopic value with time. Contrariwise, the asymptotic transverse dispersion coefficient, D_T, was found to be of the order of the local dispersion coefficient (i.e. microscopically small).

However, numerical simulations and (field and laboratory) experiments have showed an enhancement of transverse dispersion, [i.e., *Silliman and Simpson*, 1987, *Burr and Sudicky*, 1994, *Cortis and Berkowitz*, 2004]. Actually, it is widely accepted, based on empirical evidence, that the ratio between longitudinal and the transverse coefficients should be about $D_T \approx 0.1 D_L$.

Temporal fluctuations of the flow in a heterogeneous porous media were recognized as

a cause of enhanced solute spreading by *Kinzelbach and Ackerer*, 1986, and in a stochastic modelling framework by *Rehfeldt and Gelhar*, 1992. Recently, the effective spreading and mixing in time fluctuating flow through a heterogeneous medium has been studied in terms of effective dispersion coefficients [*Dentz and Carrera*, 2003, 2005; *Cirpka and Attinger*, 2003]. *Cirpka*, 2005 analyzed the transverse dispersion coefficient considering a spatially uniform flow field of a kinetically sorbing compound under sinusoidal temporal fluctuations.

Here we study the impact of the interaction of local dispersion, chemical heterogeneity and transverse temporal flow fluctuations on the temporal behaviour of the transverse effective dispersion coefficients in d=2 spatial dimensions. The chemical heterogeneity is characterized by variations in the instantaneous equilibrium sorption properties, since we account for linear instantaneous equilibrium processes.

BASICS

We define the observables that characterize solute spreading and mixing and present the transport model under consideration. Furthermore, we present the stochastic modelling approach used in this work in order to systematically integrate the spatio-temporal fluctuations of the system parameters into an upscaled transport picture and lay out the perturbation method to solve the resulting stochastic transport problem.

Observables

To quantify the influence of space fluctuations of the retardation factor and time fluctuations of the flow velocity on the effective transport behaviour, we study the effective centre of mass velocity and effective dispersion coefficients. In a stochastic model these observables are defined as averages over all typical realizations of the stochastic processes under consideration.

We define the effective centre of mass velocity and the effective dispersion coefficient as a combination of ensemble averages of the time derivatives of first and second centred moments of the (normalised) non-adsorbed solute concentration in one typical realization of the two random processes:

$$u_i^{\mathrm{eff}}(t) = \overline{\left\langle u_j(t) \right\rangle} = \frac{\mathrm{d}}{\mathrm{d}t} \overline{\left\langle m_j^{(1)}(t) \right\rangle} \tag{1}$$

$$D_{ij}^{eff}(t) = \frac{1}{2} \frac{d}{dt} \left\langle \overline{m_{ij}^{(2)}(t)} - \overline{m_i^{(1)}(t) m_j^{(1)}(t)} \right\rangle \tag{2}$$

where $m_j^{(1)}(t)$ and $m_{ij}^{(2)}(t)$ are the first two moments of the normalized spatial concentration distribution. The overbar denotes the average over the spatial random field, the angular brackets denote the average over the temporal random process. This effective quantity characterizes physical spreading in a typical disorder realization as opposed to the frequently considered ensemble dispersion coefficients, which are derived from the ensemble averaged concentration distribution as outlined in *Kitanidis*, 1988, for example.

Transport Model

The time evolution of a mobile reactive solute $c_m(\mathbf{x},t)$, with instantaneous chemical adsorption in a homogeneous porous medium in the presence of transverse temporal fluctuations of the flow is given by,

$$R(\mathbf{x})\frac{\partial c_m(\mathbf{x},t)}{\partial t}+[q\hat{e}_1+q'(t)\hat{e}_2]\cdot\nabla c_m(\mathbf{x},t)-\nabla\mathbf{D}_0\nabla c_m(\mathbf{x},t)=\delta(\mathbf{x})\delta(t). \tag{3}$$

where we define the retardation coefficient $R(\mathbf{x})$ in terms of the porosity $\phi(\mathbf{x})$ and the spatially varying distribution coefficient, $k_d(\mathbf{x})$ as $R(\mathbf{x})\equiv\phi(\mathbf{x})+[1-\phi(\mathbf{x})]k_d(\mathbf{x})$. The flow field is characterized by a constant component q in 1-direction and a temporally fluctuating component $q'(t)$ in 2-direction, $\hat{e}_i$ are the directional unit vectors. By construction $\langle q'(t)\rangle=0$. The (constant) local dispersion tensor is denoted by $\mathbf{D}_0=D_0\mathbf{I}$. The right side of (3) accounts for transport of a solute evolving from a point-like instantaneous injection. We assume vanishing concentration at the boundaries at infinity.

In a stochastic modelling framework the fluctuating retardation factor $R(\mathbf{x})$, as well as the flow velocity $q'(t)$, are represented as random processes. We split $R(\mathbf{x})=\overline{R}\left[1-\mu(\mathbf{x})\right]$ where $\overline{R}$ is the ensemble averaged retardation factor. The field $\mu(\mathbf{x})$ denote the random fluctuations around these mean values. By construction, $\overline{\mu(\mathbf{x})}=0$.

Reescaling the remaining quantities in the transport equation according to:

$$\mathbf{D}=\frac{\mathbf{D}_0}{\overline{R}} \tag{4}$$

$$g(\mathbf{x},t)=\overline{R}c_m(\mathbf{x},t) \tag{5}$$

$$\mathbf{u}(t)=\frac{\mathbf{q}(t)}{\overline{R}}=u\left[\hat{e}_1-\nu(t)\hat{e}_2\right] \tag{6}$$

The transport equation then reads:

$$\frac{d}{dt}g(\mathbf{x},t)+\mathbf{u}(t)\nabla g(\mathbf{x},t)-\nabla\mathbf{D}\cdot\nabla g(\mathbf{x},t)=\delta(\mathbf{x})\delta(t)+\mu(\mathbf{x})\frac{d}{dt}g(\mathbf{x},t) \tag{7}$$

The auto correlation functions of $\mu(\mathbf{x})$ and $\nu(t)$ are denoted by $\overline{\mu(\mathbf{x})\mu(\mathbf{x}')}=\sigma^2_{\mu\mu}C^{\mu\mu}(\mathbf{x}-\mathbf{x}')$ and $\overline{\nu(t)\nu(t')}=\sigma^2_{\nu\nu}C^{uu}(t-t')$. Both correlations are assumed to be short range, i.e., to decrease quickly for distances and times larger than the correlation length and time, respectively.

Reformulating equation (7) in terms of an equivalent integral equation in Fourier space we obtain,

$$\tilde{g}(\mathbf{k},t)=\tilde{g}_0(\mathbf{k},t,0)+\int_{k'}\int_{-\infty}^{\infty}\mathrm{d}t'\,\tilde{g}_0(\mathbf{k},t,t')\mu(k')\partial_{t'}\tilde{g}(\mathbf{k}-\mathbf{k}',t') \tag{8}$$

where we use the following short-hand notation

$$\int_{k} \ldots = \int \frac{d^{d}k}{(2\pi)^{d}} \ldots \tag{9}$$

The Fourier transformed Green function of the transport problem (7) for $\mu(\mathbf{x}) = 0$, is denoted by $\tilde{g}_0(\mathbf{k}, t, t')$ and reads as

$$\tilde{g}_0(\mathbf{k}, t, t') = \exp\left[-\mathbf{k}^T \mathbf{D} \mathbf{k} (t - t') + \mathrm{i}\mathbf{k} \cdot \int_{t'}^{t} \mathrm{d}\tau \mathbf{u}(\tau) \right] \Theta(t - t') \tag{10}$$

with $\Theta(t)$ the Heaviside step function.

Iterating the integral (8), up to the second order in terms of $\mu(\mathbf{k})$ we obtain a perturbation series.

$$\begin{aligned} \tilde{g}(\mathbf{k}, t) = \tilde{g}_0(\mathbf{k}, t, 0) &+ \int_{k'} \int_{-\infty}^{\infty} dt' \tilde{g}_0(\mathbf{k}, t, t') \mu(\mathbf{k}') \partial_{t'} \tilde{g}_0(\mathbf{k}, t', 0) \\ &+ \int_{k'} \int_{-\infty}^{\infty} dt' \int_{k''} \int_{-\infty}^{\infty} dt'' \tilde{g}_0(\mathbf{k}, t, t') \mu(\mathbf{k}') \partial_{t'} \tilde{g}_0(\mathbf{k} - \mathbf{k}', t', t'') \\ &\times \mu(\mathbf{k}'') \partial_{t''} \tilde{g}_0(\mathbf{k} - \mathbf{k}' - \mathbf{k}'', t'', 0) + \ldots \end{aligned} \tag{11}$$

This series constitutes the basis of the perturbational treatment of the transport parameters.

Transport Coefficients and time scales.

The effective dispersion coefficient $D_{ij}^{\mathrm{eff}}(t)$ provided by (2), can be written in terms of the Fourier transform of the normalized concentration distribution, $\tilde{p}(\mathbf{k}, t) = \tilde{g}(\mathbf{k}, t) / \tilde{g}(0, t)$, as,

$$D_{ij}^{\mathrm{eff}}(t) = \frac{1}{2} \frac{\mathrm{d}}{\mathrm{dt}} \left(-\mathrm{i}\partial_{\mathrm{k_j}} \right) \left(-\mathrm{i}\partial_{\mathrm{k_i}} \right) \overline{\left\langle \ln \tilde{p}(\mathbf{k}, t) \right\rangle} \Big|_{\mathbf{k}=0} \tag{12}$$

Inserting the perturbation series (11), into (12) and expanding the resulting expression up to the second order in the fluctuations of the random fields, the transverse effective dispersion coefficient is given by,

$$D_{ij}^{\mathrm{eff}} = D_{ij} + \delta^{\mu\mu} D_{ij}^{\mathrm{eff}} + \delta^{\mu\nu} D_{ij}^{\mathrm{eff}} \tag{13}$$

Equation (13) is the sum of the contributions due to local dispersion D_{ij}, chemical heterogeneity $\delta^{\mu\mu} D_{ij}^{\mathrm{eff}}$, expressions already provided by *Attinger et al*, 1999, and the interaction between transverse temporal fluctuations of the flow conditions and chemical heterogeneity $\delta^{\mu\nu} D_{ij}^{\mathrm{eff}}$. Here we focus in the transverse contribution of the dispersion coefficient $\delta_{i2}\delta_{j2} D_{ij}^{\mathrm{eff}} = \delta^{\mu\nu} D_{22}^{\mathrm{eff}}$.

The following scales determine the characteristic temporal behaviour of the effective dispersion coefficients,

$$\tau_u = \frac{l_1}{u} \tag{14}$$

$$\tau_{D_2} = \frac{l_2^2}{D_{22}} \tag{15}$$

The advection time scale τ_u measures the time for a solute particle to travel by advection a distance equal to one longitudinal correlation length l_1. The dispersion time scale τ_D is the characteristic time for dispersive solute transport over one correlation length l_2.

The ratio between these scales is $\varepsilon = \tau_u / \tau_{D_2} = D_{22} l_1 / (\bar{u} l_2^2)$ the inverse of the microscopic Peclet number. In most real situations transport is dominated by advection. Hence one expects $\varepsilon \ll 1$.

We furthermore define the non-dimensional Kubo number, $\kappa = \tau / \tau_u$, which compares the correlation time of the flow fluctuations τ, to the advection time scale τ_u. It equivalently compares the distance $l_k = \bar{u}\,\tau$, (Kubo distance) over which the solute is advected by the mean flow during one correlation time τ, to the correlation length in direction of the mean flow l_1 ($\kappa = l_\kappa / l_1$).

Note that for times smaller than the advection time scale, $t \le \tau_u$, the solute has been transported over a distance shorter than the correlation length l_1 of the medium, and has spread by local dispersion over a distance much smaller than the corresponding correlation distance. On these short scales, the medium looks homogeneous and the solute does not "see" the heterogeneity of the medium. Therefore, we concentrate on the more relevant time regime $t \gg \tau_u$ and apply the small inverse Peclet number $\varepsilon \ll 1$ approximation.

We derive analytical results for the contribution of $\delta^{\mu\nu} D_{22}^{\text{eff}}(t)$ in d=2 spatial dimensions, considering an isotropic scenario $l = l_1 = l_2$ and a point like injection of tracer in $t = 0$.

In the limit of small ε, we obtain for the contributions to the transverse effective dispersion coefficients the following expressions,

$$\delta^{\mu\nu} D_{22}^{\text{eff}}(t) = \sigma_{\mu\mu}^2 \sigma_{\nu\nu}^2 u l \left\{ M_{22}(t, \mathbf{I}) - M_{22}\left[t, \mathbf{A}(t)\right] \right\} \tag{16}$$

where the auxiliary functions $M_{22}(t, \mathbf{B})$ are defined by,

$$M_{22}(t, \mathbf{B}) = \int_{k'} \int_0^{t/\tau_u} dt' \Gamma^{\mu\mu}(\mathbf{k}') \exp\left(-\frac{k_l'^2}{2} (B_l - 1) - \mathrm{i} k_1' t' \right) C_{22}(t - t') \tag{17}$$

Where we sum over identical indices. The components of the vector $\mathbf{A}(t)$ are defined by $\mathbf{A}(t)\mathbf{I}$, where,

$$A(t) = 1 + 4\frac{t}{\tau_D} \tag{18}$$

For compactness we define the correlation function $\Gamma^{\mu\mu}(\mathbf{k}') \equiv C^{\mu\mu}(k_1'/l, k_2'/l)$.

Explicit Results

To derive explicit results, we have to specify the spatial and temporal correlation functions. A convenient choice made in the literature is Gauss-shaped functions. We use an autocorrelation function for the retardation random field $\mu(\mathbf{k}')$ which in Fourier space in dimensions reads:

$$C^{\mu\mu}(k) = (2\pi)\prod_{i=1}^{2} l \exp\left(-\frac{1}{2}(k_i\, l)^2\right) \tag{19}$$

The length scale l is the correlation length of the retardation fields. We also assume a Gaussian shaped correlation function for the temporal fluctuations of the random flow field (*Dentz and Carrera*, 2005), where the correlation function $C^{vv}(t)$ decays quickly for times that are large compared to the correlation time scale τ.

$$C^{vv}(t) = \exp\left(-\frac{t^2}{2\tau^2}\right) \tag{20}$$

Inserting the correlation functions (19) and (20) in equation (16) we obtain analytical expressions applying the approximations $\varepsilon \ll 1$ and $t \gg \tau_u$ in d=2 spatial dimension.

$$\delta^{\mu\nu} D_{22}^{\text{eff}} = \sqrt{\frac{\pi}{2}}\, \sigma_{vv}^2\, \sigma_{\mu\mu}^2\, u\, l\, \kappa \left[\frac{1}{\sqrt{1+\kappa^2}} - \frac{1}{\sqrt{\frac{4t}{\tau_D}+1}\sqrt{\frac{4t}{\tau_D}+1+\kappa^2}} \right] \tag{21}$$

DISCUSSION

Asymptotic behaviour for large times.

We consider the asymptotic behaviour of the contributions to the effective dispersion coefficients as a function of the Kubo number κ,

$$\lim_{t\to\infty} \delta^{\mu\nu} D_{22}^{\text{eff}}(t) = \delta^{\mu\nu} D_{22}^{\infty}(\kappa) \tag{22}$$

Figure 1 illustrates the asymptotic behaviour of $\delta^{\mu\nu} D_{22}^{\infty}(\kappa)$ in $d = 2$ dimensions for $\varepsilon = 0$. Analyzing transverse asymptotic contributions to the effective dispersion coefficients, we observe that in the limit $\kappa \to 0$, $\delta^{\mu\nu} D_{22}^{\infty}(\kappa)$ tends to zero. The correlation time of the velocity fluctuations is much smaller than the advection time, $\tau \ll \tau_u$. In this limit the Kubo length is much smaller than the correlation length, $l_\kappa \ll l_1$. Thus the medium "looks" homogeneous. The transverse contribution increases more or less linearly with κ for $10^{-2} < \kappa < 1$ since,

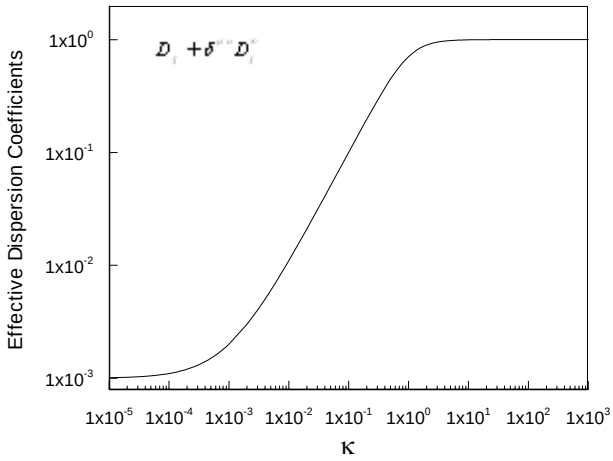

Fig. 1 Behaviour of the second order contributions to the transverse asymptotic dispersion (scaled by $\sqrt{\frac{\pi}{2}}\,\sigma_{\mu\mu}^2\,\sigma_{\nu\nu}^2 u\,l$) as a function of the Kubo number ($\kappa = \tau/\tau_u$) in $d = 2$ for $\varepsilon = 0$ and $\tau_D = 10^3\,\tau_u$.

$$\delta^{\mu\nu} D_{22}^{\infty}(\kappa) = \sqrt{\frac{\pi}{2}}\,\sigma_{\mu\mu}^2\,\sigma_{\nu\nu}^2\,u\,l\,\frac{\kappa}{\sqrt{1+\kappa^2}} \tag{23}$$

it tends to its asymptotic value of about $\sqrt{\frac{\pi}{2}}\,\sigma_{\mu\mu}^2\,\sigma_{\nu\nu}^2\,u\,l$ in the limit $\kappa \to \infty$, but it is close to this limit already for $\kappa \approx 10$.

Time behaviour.

We analyse the temporal behaviour of the transverse effective dispersion coefficients for different Kubo numbers in $d = 2$ dimensions.
Figure 2 illustrates the time evolution of the approximation for $\varepsilon = 0$, and $t \gg \tau_u$. The

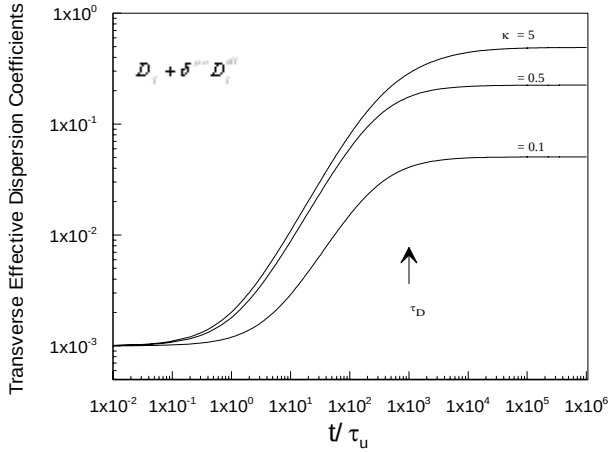

Fig. 1 Time behaviour of the contributions to the transverse effective dispersion (scaled by $\sqrt{\frac{\pi}{2}}\,\sigma_{\mu\mu}^2\,\sigma_{\nu\nu}^2 u\,l$) in $d = 2$ for $\varepsilon = 0$. $\tau_D = 1000\,\tau_u$, $\kappa = 0.1$, $\kappa = 0.5$ and $\kappa = 5$.

dispersion time scale is $\tau_D = 10^3\,\tau_u$. The advection and dispersion time scales are clearly separated. From the approximate expression given in equation (21), we identify

the Kubo time scale $\tau_\kappa = (1+\kappa^2)\tau_D = (l_1^2 + l_\kappa^2)\tau_D$, (*Dentz and Carrera*, 2005) , which measures the time for the local dispersive spreading over an effective length that is given by the correlation length l and the Kubo length $l_\kappa = u\tau$. This new scale, together with the advection scale τ_u and the dispersion time scale τ_D, separates three different time regimes: short-time regime $t \ll \tau_u$, intermediate time regime $\tau_D \ll t \ll \tau_\kappa$, and long-time regime $t \gg \tau_\kappa$.

As discussed previously, for short-times $t \ll \tau_u$, the solute plume has not yet been advectively transported over one correlation length of the medium and thus, the solute "see" the medium homogeneous. Hence, the effective dispersion coefficients remains of the order of local dispersion.

In the intermediate time regime $\tau_D \ll t \ll \tau_\kappa$ for $\kappa \gg 1$, the transverse effective coefficient evolve approximately as $\sim t^{-1/2}$, which is identical to the behaviour observed for the longitudinal component under steady state conditions, *Attinger et al*, 1999. In this time regime the flow velocity is practically stationary as the temporal fluctuation scale is much larger than the travel time $\tau \gg t$. It is equivalent to consider $\nu(t) \sim$ constant , i.e., there is a constant component in transverse direction. Thus the principal direction of the velocity flow is given by $u\hat{e}_1 + \nu_2\hat{e}_2$. Along both directions we observe macroscopic spreading, as obtained by *Attinger et al*, 1999, for the longitudinal effective dispersion coefficient in steady state conditions due to the interaction of local dispersion and chemical heterogeneities of the medium.

For times $t \gg \tau_\kappa$, the asymptotic long time value of the effective transverse dispersion is approached according to $\sim t^{-1}$. In this time regime, the travel time is much larger than the correlation time $t \gg \tau$ and the plume has been spread out over a distance larger than the Kubo length l_κ. Hence we have an interaction between transverse temporal fluctuations and chemical heterogeneities of the medium which leads to a faster increase of the transverse coefficient towards its asymptotic value.

For $\kappa \leq 1$, the asymptotic long-time regime is reached according to $\sim t^{-1}$ already for $t \gg \tau_D$ because $\tau_D \sim \tau_\kappa$, therefore we observe the same behaviour described above for $t \gg \tau_\kappa$.

REFERENCES

Attinger, S., Dentz, M., Kinzelbach, H., and Kinzelbach, W., (1999), Temporal behaviour of a solute cloud in a chemically heterogeneous porous medium, *J. Fluid Mech.*, **386**, 77-104.

Burr, D.T., and Naff, R.L., (1994), Nonreactive and reactive solute transport in three-dimensional heterogeneous porous media: Mean displacement, plume spreading, and uncertainty, *Water Resour. Res.*, 30(3), 791-815.

Cirpka, O. A., Attinger, S., (2003), Effective dispersion in heterogeneous media under random transient flow conditions,*Water Resour. Res.*, **39**(9), 1257, doi:10.1029/2002WR001931.

Cirpka, O. A., (2005), Effects of sorption on transverse mixing in transient flows,*J. Cont. Hydrol.*, **78**, 207-229, doi:10.1016/j.jconhyd.2005.05.008.

Cortis, A., and Berkowitz, B., (2004), Anomalous transport in "classical" soil and sand columns, *Soil Science Society of America Journal*, **68**, 1539-1548.

Dentz, M., Kinzelbach, H., Attinger, S., and Kinzelbach, W.,(2000a), Temporal behavior of a solute cloud in a heterogeneous porous medium: 1. Point-like injection, *Water Resour. Res.*, **36**(12), 3591-3604.

Dentz, M., Kinzelbach, H., Attinger, S., and Kinzelbach, W.,(2000b), Temporal behavior of a solute cloud in a

heterogeneous porous medium: 2. Spatially extended injection, *Water Resour. Res.*, **36**(12), 3605-3614.

Dentz, M., and Carrera, J., (2003), Effective dispersion in temporally fluctuating flow through a heterogeneous medium. *Phys. Rev.*, **E 68**, doi:10.1103/PhysRevE.68.036310.

Dentz, M., and J. Carrera (2005), Effective solute transport in temporally fluctuating flow through heterogeneous media, *Water Resour. Res.*, **41**, W08414, doi:10.1029/2004WR003571.

Kinzelbach, W., Ackerer, P., (1986), Modélisation du transport de contaminant dans un champ d'écoulement non-permanent, *Hydrolgéologie*, **2**, 197-205.

Kitanidis, P. K.,(1988), Prediction by the method of moments of transport in a heterogeneous formation, J. Hydrology; **102**; 453-473.

Rehfeldt, K.R., and Gelhar, L.W., (1992), Stochastic analysis of dispersion in usteady flow in heterogeneous aquifers, *Water Resour.Res.*, 28(6), 2085-2099.

Silliman, S. E., and Simpson, E. S., (1987), Laboratory evidence of the scale effect in dispersion of solutes in porous media, *Water Resour.Res.*, **23**, 1667-1673.